

**A Novel Analytical Method for the Determination of Residual Moisture in
Plutonium Dioxide :
Supercritical Fluid Extraction/Fourier Transform Infrared Spectroscopy**

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Abstract

A novel approach has been developed at the Los Alamos National Laboratory for the quantitative determination of moisture content in impure plutonium oxide. The method combines a commercial supercritical fluid extraction instrument using supercritical carbon dioxide (SCCO₂) with on-line detection using a high-pressure Fourier Transform Infrared Spectroscopy (FTIR) cell. The combined SCCO₂/FTIR system has been modified for use inside a fully enclosed glovebox.

A series of validation experiments were performed using a pure, surrogate oxide (ThO₂) and an inorganic hydrate (CaSO₄·2H₂O). The level of agreement between LOI and SCCO₂/FTIR for the surrogate oxide is excellent. The results for the inorganic hydrate showed excellent correlation with the known amount of water present. Results obtained for a group of nominally pure PuO₂ samples were verified by independent measurement.

The results of SCCO₂/FTIR for impure PuO₂ samples is consistently lower than the results of obtained from the current analytical method (Loss On Ignition), indicating that the current method is inadequate for analytical purposes. While further verification experiments of the SCCO₂/FTIR method are underway, these initial results suggest that SCCO₂/FTIR could be used as an alternative analytical method for the Materials Identification and Surveillance program.

Introduction

The downsizing of the nuclear weapons stockpile has generated a large amount of excess plutonium metal. The preferred form for long-term storage is as calcined plutonium dioxide (PuO₂) powder. The long-term storage of PuO₂ powder in sealed containers requires, however, that residual moisture be reduced to very low levels, to prevent the possible build-up of overpressures through radiolysis of water or other hydrogen-bearing (hydrogenous) compounds.[1] The current Department of Energy (DOE) standard method for thermal stabilization [2] specifies calcination at 950°C for two hours. Thermal stabilization (removal of

hydrogenous compounds) is verified by a Loss-On-Ignition (LOI) test, where the total weight loss following heating to 1000°C is determined. A weight loss in excess of 0.5 mass % requires that the material be re-calcined. Unfortunately, much of the PuO₂ destined for storage is impure, containing large amounts of inorganic salts. Since the LOI test relies on a simple weight difference, it cannot discriminate between weight loss due to hydrogenous compounds and volatilized, non-hydrogenous compounds, including inorganic salts. Supercritical fluid CO₂ extraction (SCCO₂) was identified as an alternative analytical method since it is a low-temperature process ($T_{\text{max}} = 140^{\circ}\text{C}$) and because CO₂, as the extraction solvent, does not readily solubilize inorganic materials and is inert with respect to reactive components which may be present in the impure oxides.

In order to evaluate SCCO₂ as a moisture analysis method, we modified a commercial extraction instrument for glovebox operation. Further, we interfaced this extraction instrument to a high-pressure FTIR detection system for quantitative detection of the extracted water. Experiments on non-radioactive matrices, including free-water, hydrates, and a high-purity, surrogate oxide, were first studied. Excellent results were obtained for the dehydration of free and hydrated water, and a high level of agreement was found between the SCCO₂/FTIR and LOI measurements for high-purity surrogate and plutonium oxides. Subsequent work on impure plutonium oxides, however, showed a significant divergence between SCCO₂/FTIR and LOI. Our results indicate that (1) the LOI method is not a reliable indicator for the moisture content of impure PuO₂, and (2) SCCO₂/FTIR is a reliable, alternative analytical method. Pending the results of a final set of validation experiments, the updated, provisional DOE standard method will include SCCO₂/FTIR as an acceptable analytical method for verification of thermal stabilization. In this paper, we present the results of the experimental program thus far and describe the final set of validation experiments.

Development

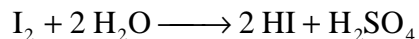
A. Supercritical Fluids

The temperature at which the vapor pressure above a pure liquid reaches one atmosphere is known as the normal boiling point. For water, the normal boiling point at one atmosphere is 100°C. In an open container, the temperature of liquid water cannot be raised above 100°C since this would cause the vapor pressure of water to exceed the ambient pressure conditions (one atmosphere). If we place a quantity of water in a *sealed* container, however, then we may heat the liquid water to temperatures above 100°C, since the vapor pressure can now rise to whatever value is in equilibrium at that temperature. As we uniformly heat this sealed container, the density of the liquid water decreases through thermal expansion. Simultaneously, the density of the water vapor increases. We can continue this heating process until the density of the liquid has been so reduced,

and the density of the water vapor has been so increased, that the two densities become equal. The temperature at which this occurs is called the critical temperature. Since the temperature inside our sealed container is everywhere equal, and the density is everywhere equal, thermodynamics requires that the pressure inside the container be everywhere equal. This pressure is called the critical pressure. A liquid which has been brought to conditions above its critical temperature and pressure is known as a supercritical fluid.

Supercritical fluids are used as solvents in many commercial applications, including the extraction of caffeine from coffee and spices and essential oils from plants for use in perfumes and foods. The attractiveness of supercritical fluids as solvents stems from their unique combination of liquid-like and gas-like properties. To a first approximation, the solvent power of a fluid is related to its density. The high, liquid-like densities of supercritical fluids therefore allow for high solubilities. Figure 1 shows the pressure-temperature-density surface for pure CO₂. The critical point for pure CO₂ (31°C and 1072 psi) as shown in this figure the solid circle. It can be seen that relatively small changes in temperature or pressure near the critical point can result in large changes in CO₂ density. It is this tunability of density, and thus tunability of solvent power, which is the most attractive attribute of supercritical fluids. For example, Figure 3 shows the equilibrium solubility of water in CO₂, as a function of pressure at various temperatures. It is seen that the solubility increases with both pressure and temperature. The conditions used for our extractions are 3000 psi and 140°C, resulting in an equilibrium solubility of approximately 5 mole %. Also, the low, gas-like viscosity of SCCO₂, Figure 2, allows for easy penetration into porous and micro-porous matrices. Also, supercritical fluids possess high, gas-like diffusivities, providing for highly effective mass transport.

The moisture analysis of PuO₂ poses unique environmental, safety and health concerns. Specifically, the handling of radioactive materials require stringent design, certification, and operating procedures. With these issues in mind, an evaluation was made on possible analytical methods for moisture determination, with particular emphasis on their applicability to radioactive materials, and suitability for glove box operation.[3] As an example, Karl Fischer titration based on the reduction of iodine,



is a very accurate method and is probably the most widely recognized analytical procedure for water determination. However, this reaction is normally performed in a non-aqueous, liquid solvent such as methanol or ether. Aside from flammability concerns, this method would generate large volumes of hazardous, liquid radioactive waste. Also, the solvent itself would suffer radiolytic decomposition, generating hydrogen.

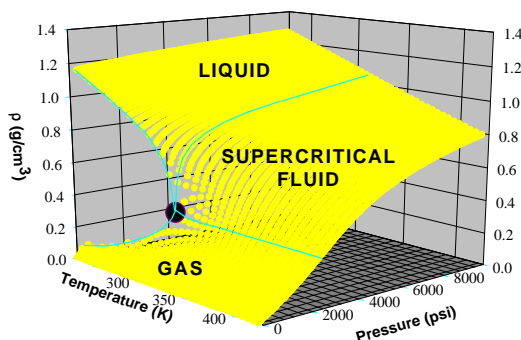


Figure 1. Density, ρ , as a function of temperature and pressure for pure CO₂.

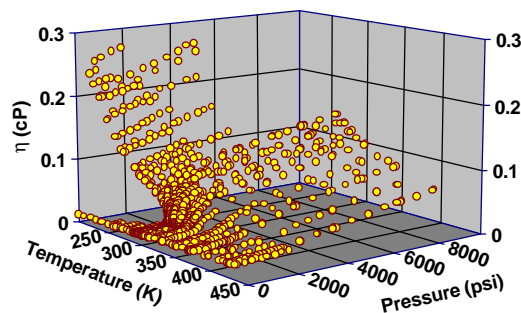


Figure 2. Viscosity, η , as a function of temperature and pressure for pure CO₂.

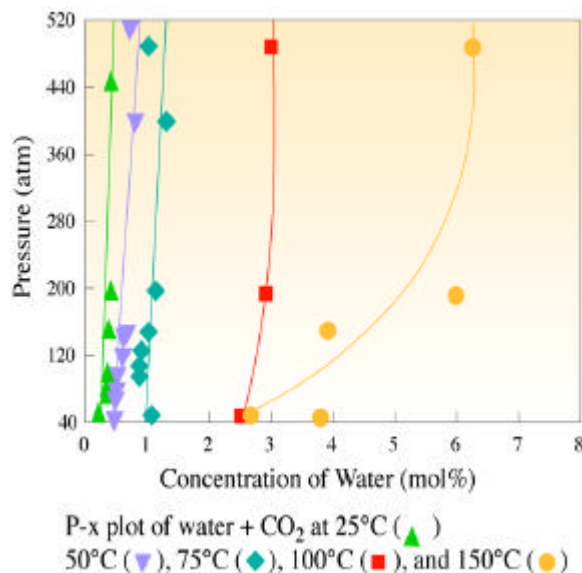


Figure 3. Equilibrium solubility of H₂O in CO₂, as a function of pressure, for various temperatures.[4]

Other analytical methods, identified through the evaluation process, are also being studied, including Interstitial Gas Analysis (IGA). This method involves combusting the sample in air, and detecting either total hydrogen or water by non-dispersive infrared (NDIR) spectroscopy. The IGA method has been used for independent comparison with SCCO₂/FTIR and LOI.

B. System Description

All PuO₂-bearing materials and *any* component that contacts these materials must be handled in a glove box to provide protection to the analyst and to the environment. Those components not requiring direct contact with the oxide material can (and should) be placed outside of the glove box, allowing for ease of maintenance and to minimize the production of radioactive waste. A schematic diagram of the SCCO₂/FTIR system, showing the placement of the major components with respect to the glovebox, is shown in Figure 4. Additional engineering modifications were made to ensure safe operation, include an inline

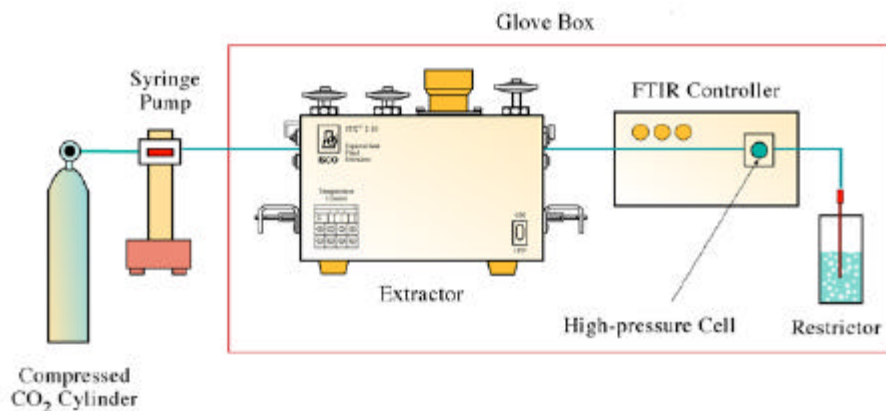


Figure 4. Schematic diagram of the SCCO₂/FTIR system.

check valve, placed directly inside the glove box, along with rigorous procedural steps, to insure the direction of the CO₂ flow is always in the direction of the glove box containment. A photograph of the actual experimental apparatus contained inside the glovebox, is shown in Figure 5. After construction of the experimental apparatus, a complete hazard evaluation was performed, based on the specific facility utility requirements of ventilation, glovebox design, and electrical power.

Supercritical fluid extraction-grade CO₂ is supplied by a helium-headspace cylinder. The supercritical extraction system is manufactured by ISCO, Inc. (Lincoln, Nebraska) and consists of three units: a Model 260D syringe pump, a model SFX-2-10 extractor and a heated, constant-flow restrictor. The detection system combines an FTIR spectrometer, manufactured by Midac Corporation (Irvine, California, Model # PROSPECT-IR) with a high pressure IR cell, with zinc sulfide (ZnS) windows (ISCO, Inc.). The combined instrumentation allows

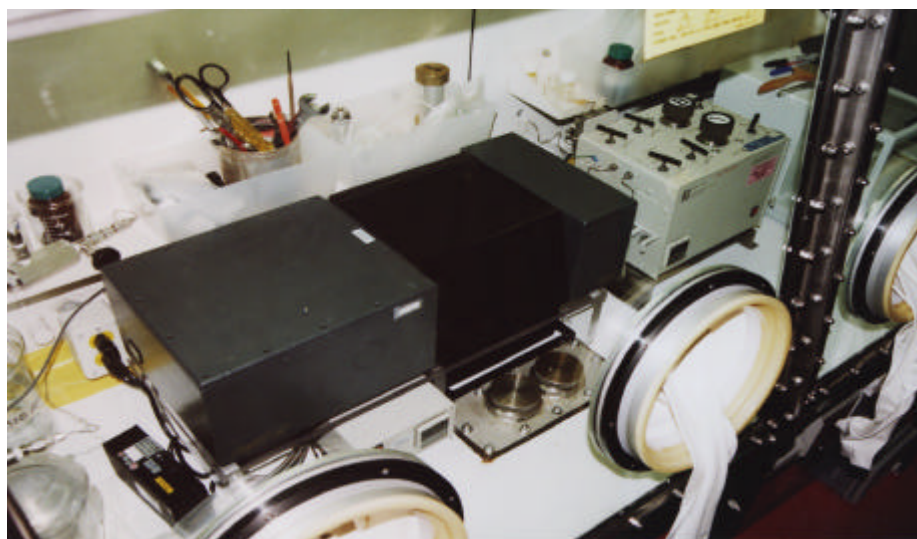


Figure 5. Photograph of the glovebox interior, showing the extractor (right) and FTIR cell (left).

for real-time monitoring during single-pass extractions. The FTIR, having a wavenumber resolution, set by the software, 1 cm^{-1} , is operated in an automatic mode where two absorption spectrums are measured every three seconds and averaged. The average is stored in a computer.

There are three infra-red active vibrational modes of the H_2O molecule : symmetric, asymmetric and scissoring of the O—H bond. While the symmetric and asymmetric modes could be used to uniquely identify the presence of water, these two modes produce broad absorption peaks occurring in the 3700 to 2900 cm^{-1} region, which would be largely obscured by absorption from the CO_2 (see the transmission spectrum in Figure 6). The absorption peak due to the scissoring mode appears in the wavenumber range 1550 - 1650 cm^{-1} . This range is well separated from CO_2 absorption and was therefore chosen for use in the quantitative analysis.

Prior to an actual extraction, an initial transmission spectrum, Figure 6, is acquired by passing pure, dry CO_2 through the FTIR cell. Next, the data collection software (AutoQuant™, Midac Corporation) is initiated and acquires a second transmission spectrum. The difference between these two transmission spectra defines the background absorption spectrum, which is used as a baseline for the extraction experiment. After the baseline is defined, the flow of CO_2 is stopped, the sample is inserted into the extractor, and the flow resumed. The data collection software now continuously monitors wavenumbers from 3800 to 500 cm^{-1} . Any water present in the sample produces an absorption peak, shown by the green curve in Figure 7. Every three seconds during the experimental run, the area of the absorption peak in the wavenumber range of 1605 to 1590 cm^{-1} (shown by the shaded green area in Figure 7) is integrated and stored in the computer. The extraction is terminated when the absorption spectrum (green curve) falls to the background spectrum (red curve). All of the stored,

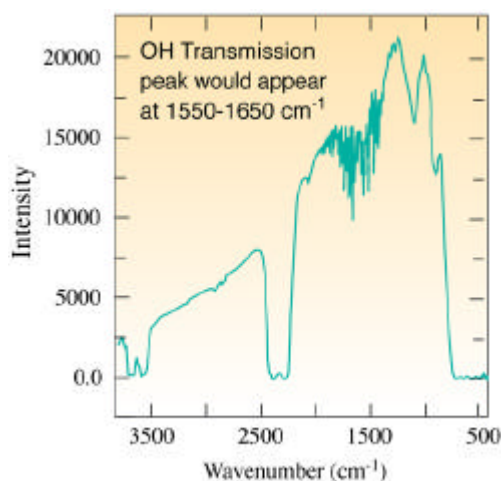


Figure 6. FTIR transmission spectra of pure, dry CO_2 . The absorption peak due to water would appear at 1550 - 1650 cm^{-1} .

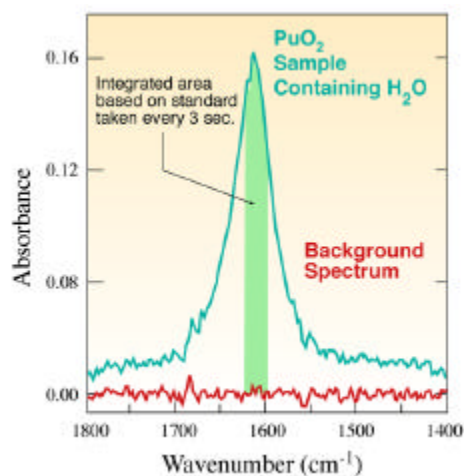


Figure 7. Absorption peak due to showing the range of wavenumbers u the integration (shaded green).

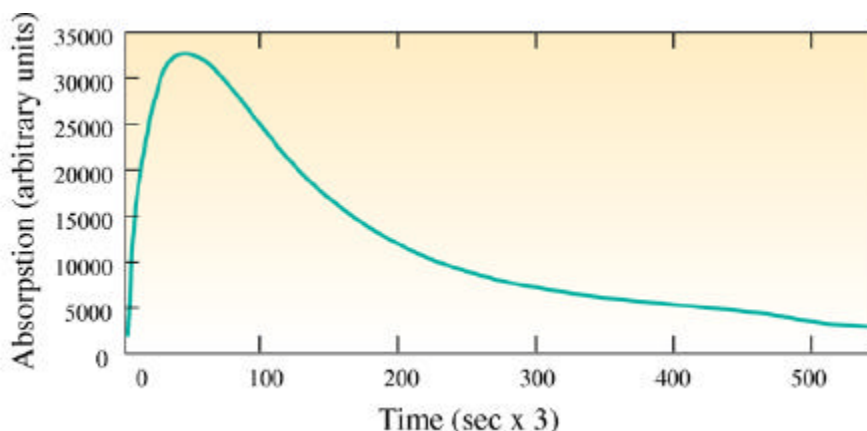


Figure 8. Typical absorption curve obtained by plotting the integrated absorption values.

integrated absorption values are then plotted against time to obtain an absorption curve, Figure 8.

Once the absorption curve is generated, its integrated area is determined. In order to correlate this integrated area to mass of extracted water, it is necessary to construct a calibration curve. This is done with a series of extractions on known amounts of liquid water, added into an extraction vessel, and gravimetrically measured with a calibrated, analytical balance. Once a calibration curve was established, Figure 9, the mass of water versus integrated area converted directly to weight percent. A Method Detection Limit (MDL) study, using the student T test at the 99% Confidence Level, is 0.08 wt. % water in a 5 g PuO_2 sample (nominal for our actual extractions). Prior to the commencement of daily analyses, a similar known-water calibration check is performed to ensure that the system is within the specified accuracy limits of $\pm 10\%$.

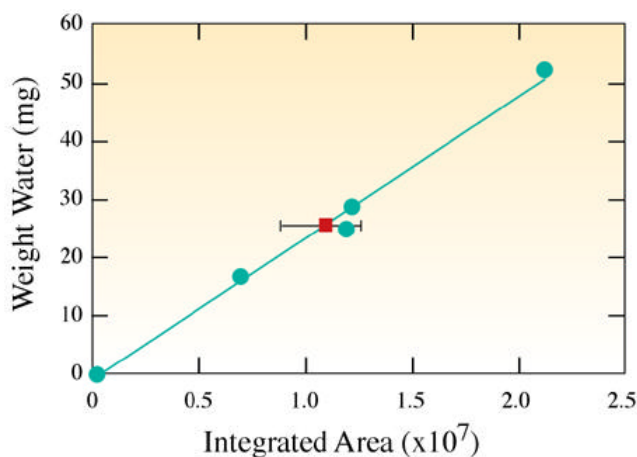


Figure 9. Experimentally measured calibration curve of free water, determined gravimetrically, versus integrated peak. The quality control guidelines (shown as a horizontal bar) indicates an acceptable calibration value, at 25 mg water (red square). This guideline is defined at 25 mg as this represents 0.5 wt. % H_2O in a 5g PuO_2 sample.

Actual analyses are made on 5 g aliquots of PuO₂, cut from a parent sample at the Plutonium Facility and placed in 10 ml internal volume PEEK cartridges which have been previously conditioned by SCCO₂ extraction at 40°C and 4000 psi. These cartridges contain 2.0 µm stainless steel frits at both ends to retain the powder within the cartridge yet allow flow of the supercritical SCCO₂. Once filled with a 5 g PuO₂ sample, the extraction cartridges are then placed inside of stainless steel or glass canisters with desiccant (Drierite™) and transferred to the facility housing the SCCO₂/FTIR system. Once transferred into the glovebox, the cartridge is removed from the canister and placed inside the extractor. The keypad-operated syringe pump controller (not shown in Figure 4), delivers the CO₂, at a constant pressure (3000 psi), to the extractor, which maintains the CO₂ and the sample at constant temperature (140°C). The transfer line, joining the extractor and the FTIR cell, has an independent temperature controller which maintains a constant temperature of 100°C. Heating of the transfer line prevents the water from condensing before entering the FTIR cell. The CO₂ is vented through a restrictor which maintains the flow rate of CO₂ through the cell at 1.0 - 1.5 ml/min. The restrictor is maintained at 90°C to avoid plugging by the formation of dry ice (adiabatic expansion) and/or precipitation of solubilized compounds. The CO₂ stream is bubbled into a dilute HNO₃ solution to retain any PuO₂ particles.

C. System Validation/Verification

Initial validation of the SCCO₂/FTIR method used a pure (99.99 %), surrogate oxide, ThO₂, where it is certain that there are no impurities other than adsorbed water. Table 1 shows the results of SCCO₂/FTIR and LOI for this material. The level of agreement is excellent.

Table 1. Results of SCCO₂/FTIR and LOI for pure ThO₂ powder.

	Wt.% H ₂ O SCCO ₂ /FTIR	Wt.% H ₂ O LOI
99.99 % pure ThO ₂	0.12	0.143
	0.12	0.157
	0.13	

A second validation was made using known amounts of water, chemically bound as calcium sulfate dihydrate (gypsum), CaSO₄·2H₂O. The results, shown in Table 2, indicate that the SCCO₂/FTIR system can successfully extract and quantitatively determine chemically bound water.

Table 2. Experimental results of SCCO₂/FTIR for CaSO₄·2H₂O.

Wt. CaSO ₄ ·2H ₂ O (mg)	Actual wt. H ₂ O (mg)	Integrated area	Calculated wt. H ₂ O (mg)
0	0	0	0
47.8	10.0	2.6986×10^6	7.7
110.0	23.0	6.2773×10^6	17.9
246.9	51.7	1.8788×10^7	53.4

Results and Discussion

We now present the results for actual samples for which weight losses have been independently measured by SCCO₂/FTIR, LOI, and IGA.

Table 3 shows comparative results for additional, nominally pure (≥ 82 wt.% Pu) PuO₂ samples. (If a sample were entirely pure PuO₂, chemical analysis would give the Pu content as 88 wt. %.) Column 2 gives the analyzed Pu content, while column 3, 4, and 5 give the measured weight losses measured by SCCO₂/FTIR, IGA and LOI, respectively. In column 6, we give the weight percentages of impurity elements, determined by separate chemical analysis, which showed large composition changes as a result of thermal stabilization at 950°C. Also in column 6 is a summation of these weight changes. This summation represents a minimum weight loss which would should be measured by LOI. It can be seen that there is, in fact, a strong correlation between the depletion of these volatile impurities and the LOI results.

All of the samples pass the 0.5 mass % limit for water content based on the SCCO₂/FTIR method. Three of four samples, the exception being CAN92, also pass the criteria according to IGA analysis. LOI analysis, however, indicates a failure of thermal stabilization for three of the four samples, and would therefore require re-calcination prior to storage. The reason for the high LOI value for the PBO sample is not known, but no elemental analysis was done on the as-received sample (prior to thermal stabilization). The other samples, however, have shown other sources for the weight lost in the LOI analysis and confirm the low levels of water seen in the SFE/FTIR and IGA analysis. For example, sample 7001 showed a 0.7% carbon content in the sample prior to calcining and, after calcining at 950°C the carbon content was essentially zero. This corresponds very well to the LOI value of 0.82% measured for this sample. Similarly, sample PPSL showed a cumulative loss of elemental impurities amounting to 0.27%, in good correspondence to the LOI value of 0.35%. Finally, CAN92 had a combined elemental impurity loss of 0.56%, with a LOI of 0.58%. The high IGA analysis for this sample can be also be related to the volatilization of elemental impurities. The analysis involves heating the sample to 400°C, which, although not as

Table 3. Comparison of experimental results for pure and nearly pure PuO₂ samples.

Sample ID	Pu (Wt. %)	wt. % H ₂ O (SCCO ₂ / FTIR)	wt. % H ₂ O (IGA)	wt. % H ₂ O (LOI)	wt. % impurity changes
					total wt. % change, Σ
7001	82	0.48	0.50	0.82	0.7% $\rightarrow \approx 0\%$ C
					$\delta \geq 0.7\%$
PPSL 365	83	< 0.08	0.01	0.35	0.02% $\rightarrow \approx 0\%$ Cl
					0.3% $\rightarrow ?$ Na
					$\delta \geq ?\%$
PBO	87	< 0.08	0.07	0.75	“nominally pure”
CAN 92	83	0.17	0.73	0.58	0.36% $\rightarrow 0.08\%$ Ga
					0.40% $\rightarrow 0.12\%$ Fe
					$\delta \geq 0.56\%$

extreme as LOI (1000°C), presents the potential for partial depletion of volatile, elemental impurities, and their interference in the IR detection.

Table 4 shows comparative results for impure (< 82 wt.% Pu) PuO₂ samples. We again perform a summation of the impurity element depletion resulting from stabilization. Again there is a strong correlation between impurity loss and LOI results. The analyses of these impure PuO₂ samples further demonstrate the trend between volatile impurity loss and LOI seen for the nominally pure samples. Also, the SCCO₂/FTIR analyses again show much lower moisture levels than LOI.

There are several analytical benefits offered by the SCCO₂/FTIR method for the characterization of PuO₂. Although we currently monitor only a small wavenumber range (1590-1605 cm⁻¹), the present system acquires the entire IR spectrum (3800 to 500 cm⁻¹). Consequently, the absorption frequencies specific to organic compounds (vibrational modes due to C-H, C-O, C=O, *etc.* bonds) can be detected simultaneously and independently. We can therefore detect not only water, but also other hydrogenous material, such as plastics. This selectivity, which is a major advantage for SCCO₂/FTIR, is not possible with LOI and would require multiple analyses by the IGA method. In addition to enhanced selectivity, there is also a significant time savings. LOI requires from 8 to 10 hours per analysis, while SCCO₂/FTIR and IGA each require 30 to 60 minutes. Such time savings is very important for production systems which require real time analysis in order to maintain high throughputs.

While the IGA method can, in principle, provide data on total hydrogen and therefore the quantity of water plus other hydrogenous material, it is limited to very small sample sizes (< 10 mg). Since the actual oxides to be analyzed are often highly heterogeneous, IGA would require multiple runs on the same sample

Table 4. Comparison of experimental results for impure PuO₂ samples.

Sample ID	Pu (Wt. %)	wt. % H ₂ O (SCCO ₂ /FTIR)	wt. % H ₂ O (IGA)	wt. % H ₂ O (LOI)	wt. % impurity changes
					total wt. % change, Σ
ARF	60	0.15	0.51	9.32	0.4% → 0.1% C 4.4% → 1.6% Na 1.1% → 0% Cl 5.7% → 0.2% K 1.0% → 0.5% Mg
					$\bar{O} \geq 10.2\%$
1407	68	0.38	1.20	7.10	4.0% → 2.5% Ni 4.0% → 0% S
					$\bar{O} \geq 5.5\%$
1589	77	0.48	0.50	4.32	0.7% → 0% C 0.4% → 0.3% Na 0.7% → 0.36% K
					$\bar{O} \geq 11.4\%$
BLO	79	0.58	1.70	2.48	0.2% → 0% C 6% → 0% Am
					$\bar{O} \geq 6.2\%$

to ensure a representative result. The SCCO₂/FTIR method analyzes large samples, up to approximately 15 g. Using a high-pressure FTIR cell, the H₂O is detected while dissolved in the high-pressure CO₂-phase, eliminating the problems associated with detection methods which operate at near-ambient pressures, such as hygrometers [5] and the IR detector used in the present IGA analyses. Also, the automated, FTIR detection system is expected to reduce systematic errors associated with manual data collection and integration, as well as provide improved minimum detection and reporting limits.

In addition to these analytical advantages, there are other factors which make the SCCO₂/FTIR method preferable to other analytical techniques. The current system produces a minimum amount of radioactive waste, which represents an important constraint imposed by Los Alamos and federal regulations. Further, the SCCO₂/FTIR system is relatively inexpensive and occupies a small footprint (area of the glovebox required for the apparatus), which is beneficial to design concerns for safety and cost considerations.

At this point, we wish to mention some caveats that we have identified during approximately one year of routine, analytical use of the SCCO₂/FTIR system. Plugging of the restrictor has occurred on several occasions. This may be due to small oxide particles escaping through the 2 μ m stainless steel frit at the

outlet end of the extraction cartridge as well as the 0.5 μm stainless steel frit at the outlet of the extractor. Alternatively, it could be due to solubilized organic or inorganic compounds that are precipitated during depressurization of the CO_2 . Monitoring of the LCD display on the syringe pump controller during a run will immediately indicate when this problem occurs, at which time the run can be stopped and the restrictor either cleaned or replaced. A second problem is fogging of the FTIR cell ZnS windows. ZnS is susceptible to acid attack, and we suspect that those samples containing a significant amount of nitrate or sulfate, converted to nitric or sulfuric acid by combination with the solubilized water, may be responsible. This problem will be detected during the daily calibration check. If an unusually high decrease in transmission is seen in the background spectrum, the windows must be replaced. This has been necessary twice during one year of actual analyses (approximately 70 PuO_2 samples). The use of other window materials is an option that we may pursue. We note that restrictor cleaning/replacement and window replacement can be performed within the glove box.

Additional experiments on pure PuO_2 samples “spiked” with known amounts of water, as $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$, as well as plastic (polyethylene), are being performed to further validate the method.

Conclusions

The level of agreement between LOI and $\text{SCCO}_2/\text{FTIR}$ for the pure ThO_2 surrogate oxide is excellent, indicating that the present analytical method is effective for the quantitative determination of moisture content in oxide powders. Also, the experiments performed on the $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$ samples shows that hydrated water, which is more energetically bound than adsorbed water, can also be removed and quantitatively determined.

The results of $\text{SCCO}_2/\text{FTIR}$ for the actual PuO_2 samples is consistently lower than LOI, indicating that heating to high temperature results in the volatilization of non-hydrogenous material. The use of the present $\text{SCCO}_2/\text{FTIR}$ method will therefore greatly reduce the number of “false positive” results, allowing a reduction in the time (and cost) associated with PuO_2 processing.

We would like to reaffirm that the primary design consideration for this and all analytical work with radioactive material is safety. The $\text{SCCO}_2/\text{FTIR}$ process development described in this paper has included rigorous safety reviews, by both Los Alamos and DOE, to ensure the process is performed in a manner that will protect the analyst, the Laboratory, and the environment.

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